



AFRL-OSR-VA-TR-2015-0064

Ultra-sensitive Absorption Diagnostics of Thin Films for High-power Laser Interference Coatings

Jorge Rocca
COLORADO STATE UNIVERSITY

01/05/2015
Final Report

DISTRIBUTION A: Distribution approved for public release.

Air Force Research Laboratory
AF Office Of Scientific Research (AFOSR)/ IOS
Arlington, Virginia 22203
Air Force Materiel Command

REPORT DOCUMENTATION PAGE				Form Approved OMB No. 0704-0188	
Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing this collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden to Department of Defense, Washington Headquarters Services, Directorate for Information Operations and Reports (0704-0188), 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number. PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ADDRESS.					
1. REPORT DATE (DD-MM-YYYY) 30/12/2014		2. REPORT TYPE Final Report		3. DATES COVERED (From - To) October 1, 2013- September 30, 2014	
4. TITLE AND SUBTITLE Ultra-sensitive Absorption Diagnostics of Thin Films for High-power Laser Interference Coatings				5a. CONTRACT NUMBER	
				5b. GRANT NUMBER FA9550-13-1-0201	
				5c. PROGRAM ELEMENT NUMBER	
6. AUTHOR(S) Jorge J. Rocca (PI) Oscar E. Martinez Carmen S. Menoni Esteban Domene				5d. PROJECT NUMBER	
				5e. TASK NUMBER	
				5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Department of Electrical & Computer Engineering, Colorado State University, Fort Collins, CO 80523-1373 Faculty of Engineering, University of Buenos Aires, Buenos Aires, Argentina				8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) DOD-USAF-Air Force				10. SPONSOR/MONITOR'S ACRONYM(S)	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S)	
12. DISTRIBUTION / AVAILABILITY STATEMENT Distribution A					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT The goal of this project was to demonstrate a simple method to measure absorption loss in thin dielectrics films, in particular in multilayer coatings for high power lasers in which very low absorption losses are required. The absorption characterization method is a photo-thermal technique based on thermal lensing. The method measures the defocusing of an incident probe beam caused by a thermal lens generated by absorption of the incident pump beam. The simplicity and versatility of the method allow it to be incorporated into a deposition chamber to measure absorption loss in situ. Excellent results were obtained with both amplitude modulated and pulsed pump laser beams. Results obtained in this project demonstrated the method can measure absorption losses below 10 ppm at 1 μ m wavelength. Future work consists of the development of a prototype suitable for incorporation into the deposition chamber. Such powerful diagnostics does not presently exist.					
15. SUBJECT TERMS Thin film coatings, absorption measurements, diagnostics of thin film coatings, high power laser coatings.					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES 11	19a. NAME OF RESPONSIBLE PERSON Jorge Rocca
a. REPORT NC	b. ABSTRACT NC	c. THIS PAGE NC			19b. TELEPHONE NUMBER (include area code) 970-491-8371

Ultra-sensitive Absorption Diagnostics of Thin Films for High-power Laser Interference Coatings

Grant No. FA9550-13-1-0201

DOD-USAF-Air Force

FINAL REPORT

December 29, 2014

Dr. Jorge G Rocca

**Electrical and Computer Engineering
Colorado State University**

Dr. Oscar E. Martínez

School of Engineering, University of Buenos Aires

Senior Personnel: Prof. Carmen Menoni

Colorado State University

Project Title: *Focus Error Thermal Lensing Spectroscopy for Absorption Characterization*

Abstract

The goal of this project was to demonstrate a simple method to measure absorption loss in thin dielectrics films, in particular in multilayer coatings for high power lasers in which very low absorption losses are required. The absorption characterization method is a photo-thermal technique based on thermal lensing. The method measures the defocusing of an incident probe beam caused by a thermal lens generated by absorption of the incident pump beam. The simplicity and versatility of the method allow it to be incorporated into a deposition chamber to measure absorption loss *in situ*. Excellent results were obtained with both amplitude modulated and pulsed pump laser beams. Results obtained in this project demonstrated the method can measure absorption losses below 10 ppm at 1 μm wavelength. Future work consists of the development of a prototype suitable for incorporation into the deposition chamber. Such powerful diagnostics does not presently exist.

1. Introduction

Interference coatings (IC) for high power lasers consist of stacks of alternating layers of dielectric materials that are essentially transparent at the laser wavelength. A criterion of performance in these coatings is the amount of absorbance at the laser wavelength. The growth process for IC has been optimized to achieve routinely absorption losses of less than 50 ppm. However, when the intracavity laser power is in the hundreds of KWs, even this low absorbance is a hinder. This is because absorption in the film can lead to thermal and mechanical instabilities. It is highly desirable to optimize the IC deposition process to realize even lower losses. Presently measurement are done *ex situ* after the coating run is done. From the processing standpoint it would be advantageous to have an instrument that could measure absorption loss during deposition. This will enable modifications of the deposition conditions while the coating run proceeds to realize IC with the lowest possible loss.

The goal of this project is to demonstrate a simple method to measure absorption loss in thin film dielectrics. The absorption characterization method is a photothermal technique based on thermal lensing. The method measures the defocusing of an incident probe beam caused by a thermal lens generated by absorption of the incident pump beam. The specific application is for thin films deposited on fused silica substrates. The method is simple and versatile which will allow it to be incorporated into a deposition chamber to measure absorption loss *in situ* as the different layers in the IC are grown.

The concept of the method is simple. An incident pump power of a wavelength selected to be that at which one desires to know the absorption is absorbed by the thin film. This absorption acts as a heat source that propagates through the substrate thus causing a temperature increase. Since the refractive index of the substrate changes with temperature ($(dn/dT)_{\text{SiO}_2} = 1.28\text{E-}5 \text{ 1/}^\circ\text{K}$) this acts as a lens focusing a probe beam of shorter wavelength that passes through the whole thickness of the substrate. Thus the

final focus of the probe beam is altered by the absorption of the sample. Quantifying the change in the focal plane of the probe beam allows the determination of the sample absorption. As shown below, results of initial experiments show the method can measure absorption losses below 10 ppm at 1 μm wavelength indirectly using a focus error (FE) signal.

Our motivation to develop a compact and simple method to measure absorption loss in IC is driven by the need to characterize the ion beam sputtering process used to grow IC. In this project the group from the University of Buenos Aires collaborated with the group of Prof. Menoni and Prof. Jorge Rocca at Colorado State University who conduct laser research and uses a Veeco Spector Ion Beam Sputtering system to grow dielectric IC for high power lasers operating in the mid-infrared. When fully developed the instrument will be incorporated into the deposition chamber to measure absorption loss *in situ* during processing.

The following sections describe the concept of the instrumentation and how it has been implemented. Results of a set of initial measurements are also described. These results are very encouraging in that they show the viability of measuring absorption in thin films deposited on fused silica substrates, achieving levels below 10 ppm @ 1 μm . The results were obtained during a 3 week visit of Dr. Domene and Dr. Martinez to Colorado State University.

2. Experimental Setup

The experimental setup consists of a thermal lens technique where the detection scheme is based on an astigmatic beam which impinges on a 4 quadrant detector and a focus error signal is generated (see figure 1 and figure 2). The pump laser is focused by a converging lens and the sample is placed after the beam waist so as to be able to adjust the pump beam size on the surface of the film and not have the focus inside the substrate (thus decreasing the thermal lens signal from the SiO_2 substrate). Two different pump lasers were used: a 20 W fiber coupled IPG laser model YLR-20-1064-LP; a Spectra Physics Quanta Ray GCR190 pulsed Nd:YAG laser (590 mJ 200 μs pulses at 10Hz repetition). The probe beam is a 10 mW He:Ne laser that is attenuated using ND filters (the level of attenuation depends on the transmission of the sample). Both beams have to overlap on the film surface. In order to achieve this, the sample is mounted on a translation stage in order to locate the surface in the correct plane (this is easily achieved by maximizing the signal). Two identical cylindrical lenses of same focal length, separated a distance D_z and rotated 90° between each other, generate an astigmatic probe beam. A four quadrant detector with its axis at 45° with respect to the cylindrical lenses is located at a distance $f+D_z/2$, the plane at which the probe has equal beam waists (circular beam).

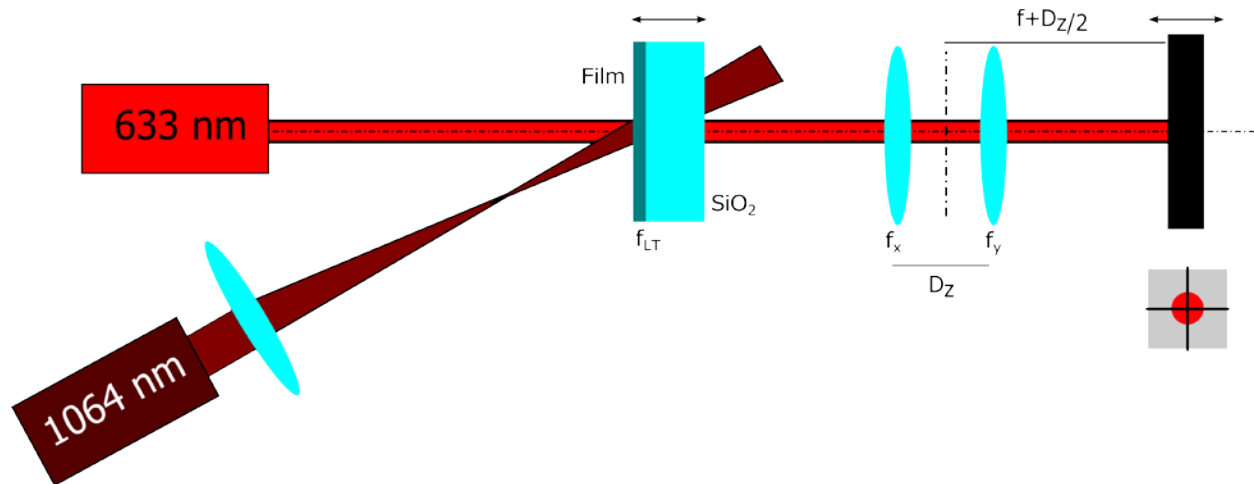


Figure 1: Experimental setup used to characterize absorption on dielectric thin films. The technique uses a 1064 nm pump laser (amplitude modulated or pulsed) and a 633 nm probe laser which overlap on the film surface. Absorption of the pump beam generates a thermal lens (f_{LT}) in the fused silica substrate which defocuses the probe beam. Two identical cylindrical lenses (focal distance $f_x=f_y=f$), rotated 90° between each other are placed at a distance of D_z of each other. Finally a four quadrant detector rotated 45° with respect to the axis of the lenses is placed at a distance $f+D_z/2$. Changes in the spatial profile of the probe beam are sensed. Both the sample and four quadrant detector are mounted on linear stages.

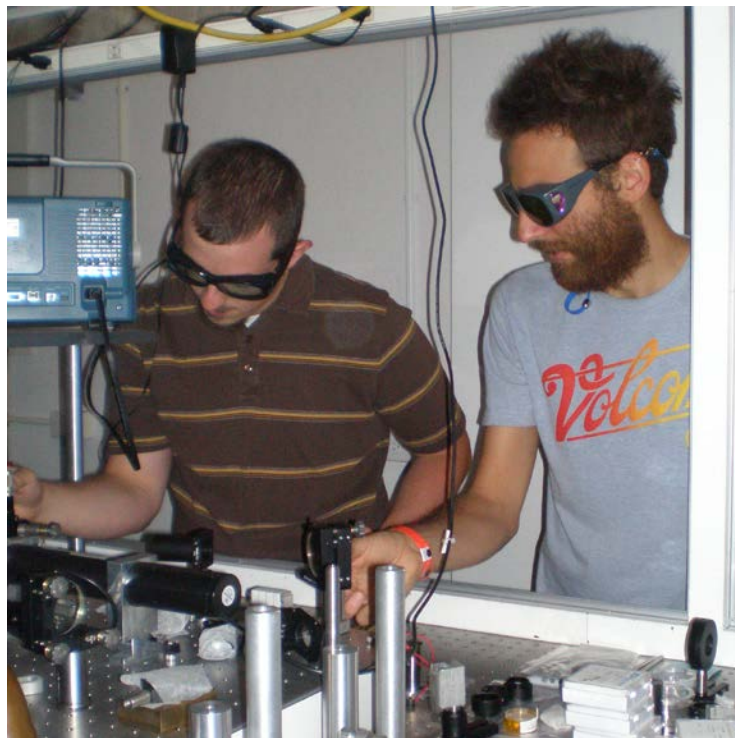


Figure 2: Graduate student Drew Schiltz (Colorado State University) and post-doctoral researcher Esteban Doméné (University of Buenos Aires) aligning the experimental setup on the optical table at CSU.

As the beam propagates, due to its acquired astigmatism, it will converge on one axis first and then on the other. The beam spatial profile as it propagates away from the lenses will first be an ellipse in one

axis, then a circle and finally an ellipse in the orthogonal axis (see figure 3). Thus by calibrating the beam spatial profile as a function of distance, it is possible to measure distances very precisely. The diagonal unbalance of the four quadrant detector generates a focus error signal (FE). The linear part of the FE signal is used to determine a thermally induced defocusing of the probe beam and thus evaluate thermal absorption of the sample. The electronics that generate the FE signal also generate the SUM signal which is the sum of all the signals from the four quadrant detector. A lock-in amplifier is used to measure the AC component of the FE signal. The analog inputs of the lock-in amplifier allow us to simultaneously monitor the DC component of the FE signal as well as the SUM. This last DC signal is used to normalize the AC FE signal and compensate for changes in probe power reaching the detector due to reflection of the sample.

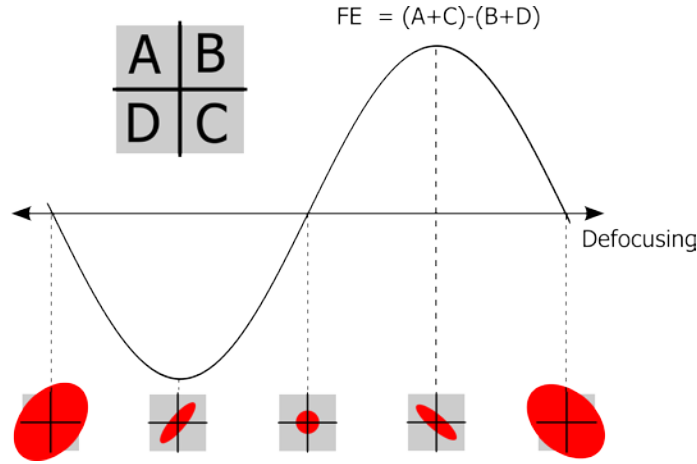


Figure 3: Propagation of astigmatic probe beam and signal detected on the 4 quadrant detector. A focus error (FE) signal is generated by measuring the unbalance between the diagonals of the detector. The linear range allows characterization of the defocusing of the probe beam.

3. Results

A. Current modulated pump laser

Using a 20 W fiber coupled IPG laser model YLR-20-1064-LP current modulated at 5Hz as the pump beam, the absorption loss of high absorbing samples was measured using two detection methods: a direct measurement of incident, transmitted and reflected optical power using a calibrated detector (scattering losses are discarded); characterizing the defocusing of the probe beam due to the thermal lens by measuring the FE signal (proportional to absorption). A comparison of both measurements can be seen in figure 4. The linear fit evidences the good correlation between measurements. Since FE signal is proportional to absorption, we use a high absorbing sample to calibrate the system. Also the measured AC FE signal is normalized by the DC SUM signal to compensate for changes in the amount of probe power reflected by different samples.

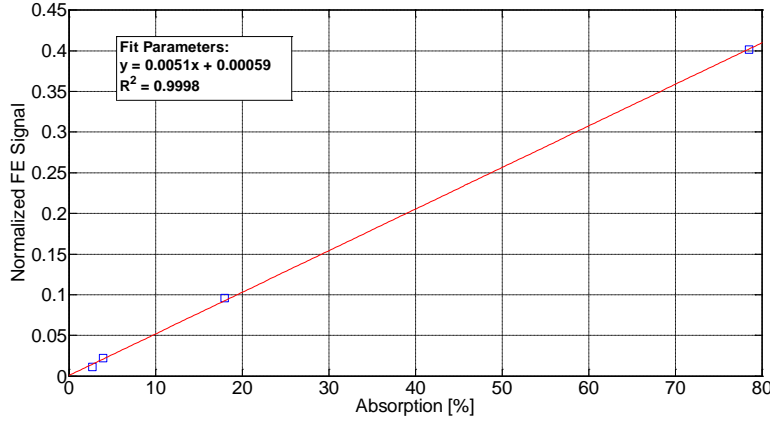


Figure 4: Comparison between absorption measurements by direct measurement of incident, reflected and transmitted pump power and by measuring AC FE signal normalized by the DC SUM signal. A linear fit of the points measured is shown with a R^2 value of 0.9998.

Samples with absorption loss as low as 12 ppm (determined by another absorption technique called PCI - Photothermal Common-Path Interferometry) were successfully measured. The signal from samples with absorption losses below 5 ppm competes with the signal from a clean fused silica substrate which undergoes bulk absorption and thus also generates a thermal lens which defocuses the probe beam. Being able to measure substrate absorption is another advantage of the technique. This allows characterization of substrates from different manufacturers.

B. Pulsed pump laser:

A Spectra Physics Quanta Ray GCR190 pulsed Nd:YAG laser is used as the pump laser to generate a time dependent thermal lens. The AC FE signal is now monitored on a digital oscilloscope triggered with the pulse trigger of the pump laser. Arrival of the pulse at the sample surface generates a thermal lens and thus the FE signal suffers a fast rise (the shorter the pulse duration the steeper the rise). Then the thermal signal inside the material decays at a slower rate. The amplitude of the rise characterizes the absorption of the sample. Figure 5 shows three different FE curves (all averaged with 512 curves) for: a Y_2O_3 thin film sample with 640 ppm absorption loss; an AR coating for 1064 nm (made up of Sc_2O_3 and SiO_2) with 113 ppm absorption loss; noise level of FE signal with pump laser off.

As in the case of a modulated pump laser, the FE signal from the oscilloscope has to be normalized by the SUM signal and then correlated with another absorption method (with a high absorbing sample). To improve sensitivity of the method, curve fitting of the FE signal can be done so as to obtain, with the intrinsic noise of the signal, the amplitude of the first rise and thus compare absorption between samples.

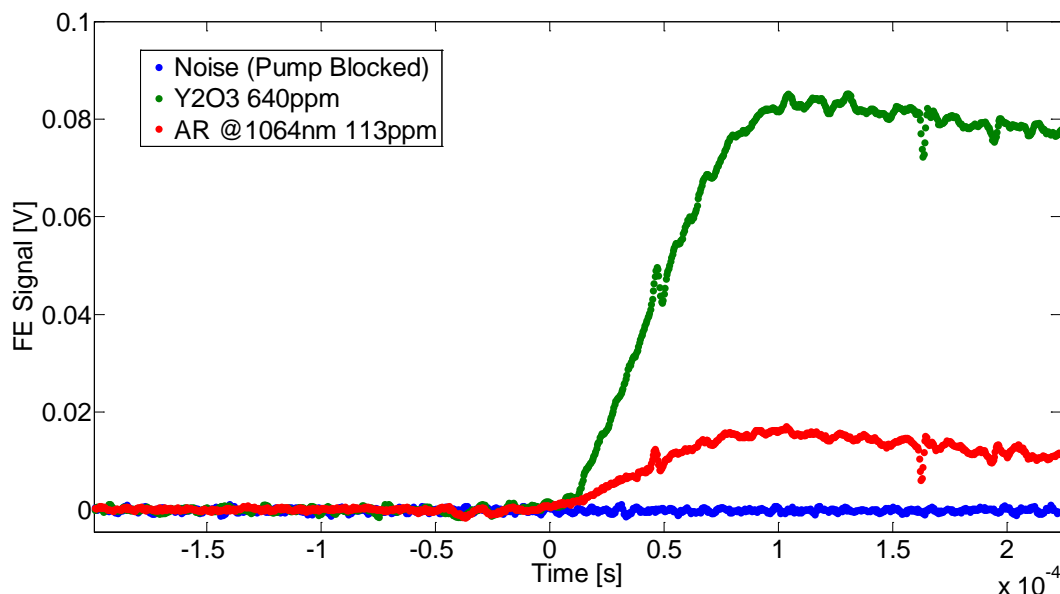


Figure 5: Averaged FE signal vs time for pulsed pump excitation (each curve is the average of 512 consecutive single shot curves). Three curves are shown: noise measurement without pump laser (blue); Y_2O_3 sample with 640 ppm absorption (green); Anti-reflective coating at 1064nm with 113 ppm absorption (red). The change in height of the first rise is due to the difference in absorption between the samples.

4. Participating personnel and exchange visits

This project has been instrumental for establishing a fluent collaboration between the School of Engineering of the University of Buenos Aires and the Department of Electrical & Computer Engineering at Colorado State University. Dr. Esteban Domené spent 3 weeks at Colorado State University setting up the thermal lensing method and testing it. In these efforts he closely collaborated with graduate student Drew Schiltz and with Prof. Menoni and Rocca. Prof. Oscar Martinez and Mr. Gustavo Sanchez visited Colorado State University for one and two weeks respectively to participate in the research.

Short course on Thin Films and Nanostructures taught by Prof. Carmen Menoni (CSU) at the University of Buenos Aires (6-12 August 2014)

During August 6-12 2014 Prof. Carmen Menoni taught a course on the fundamentals of thin film and nanostructures, a subject closely connected to this grant. Graduate students assisting the course and passing the course evaluations received credit towards their Ph.D degree.

Course title: Thin film and nanostructures: growth and characterization

Course Location: Departamento de Física. FCEyN. UBA

Number of students who attended: 9

Course Description: This course introduced students to methodologies employed in the growth of thin films and nanostructures, and to the chemical and physical mechanisms associated with the growth process and their effect in the material's characteristics. The course covered fundamentals of thin film growth.

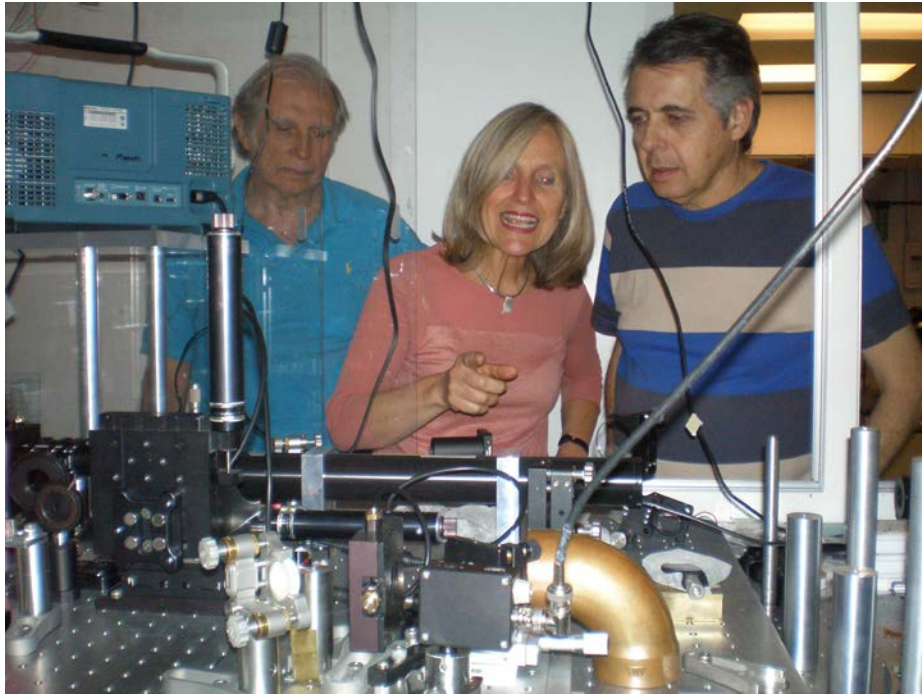
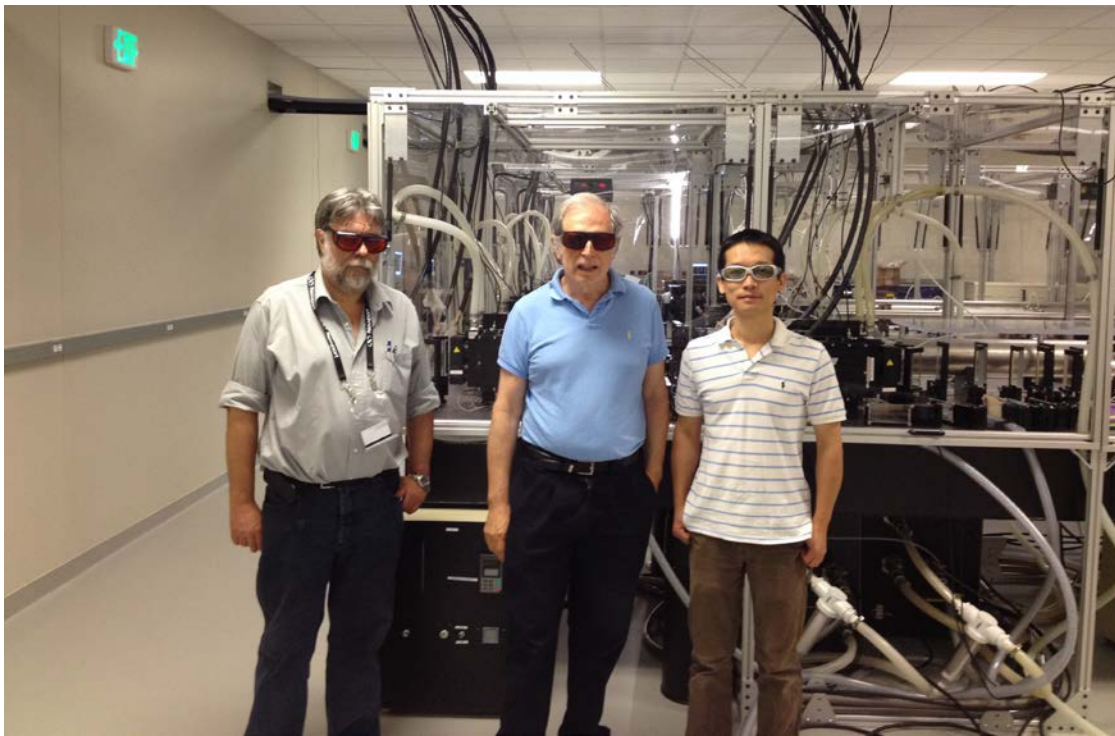


Figure 6 –Prof. Oscar Martinez discussing the experiments at Colorado State University with Prof. Jorge Rocca, and Prof. Carmen Menoni



Gustavo Sanchez (University of Buenos Aires) with Prof. Jorge Rocca and Research Scientist Dr. Yong Wang during his work visit to CSU

5. Summary

A focus error photo-thermal technique has been successfully implemented to characterize absorption loss in thin films deposited on fused silica substrates. The technique consists of a pump laser which generates a thermal lens in the sample and thus defocuses the probe laser beam. The absorption loss is obtained by measuring the defocusing of the probe beam. The technique has a sensitivity of less than 10 ppm absorption @1064nm wavelength. Sensitivity is limited by the bulk absorption of the fused silica substrate which is a few ppm. Excellent results were obtained with both amplitude modulated and pulsed pump laser beams. One of the main advantages of the technique is that the pump beam does not need to be tightly focused on the sample and the detection scheme can be placed far away from the sample (a collimated probe beam is used). These experimental advantages are essential in order to implement the system into a deposition chamber and thus allow for *in situ* measurements of absorption loss as the sequence of thin films that make up an IC are being deposited. Realizing IC with absorption losses of ~10 ppm is critical for high power laser applications where thermal and mechanical instabilities due to absorption can catastrophically damage the coating.

6. Project Participants

Graduate Students: Drew Schiltz (Colorado State University) and Gustavo Sanchez (University of Buenos Aires).

University of Buenos Aires: Dr. Esteban Domené (Post-doc), Gustavo Sanchez (Graduate Student), Prof. Oscar Martinez (PI)

Colorado State University: Drew Schiltz (Graduate Student), Dr. Dinesh Patel (Research Scientist Colorado State University), Prof. Carmen S. Menoni, Prof. Jorge J. Rocca (PI)).

7. Future Work

The next step in the project is to develop a prototype suitable for incorporation into the deposition chamber of a Veeco Spector® Ion Beam Sputtering (see figure 7a). This will allow for measurements of absorption during the fabrication of IC. Samples inside the chamber are mounted on a planetary system which rotates the samples to increase film homogeneity. This represents an experimental challenge that must be overcome. A pulsed pump laser should be used in order to be able to overlap the heating and the probe laser beams both spatially and temporally (since the sample is moving). A modulated pump laser scheme would not be suited because the heated region will be moving as the probe signal is being integrated. The proof of principle experiments using a pulsed pump laser to measure absorption, described in detail in the previous section, are very encouraging.

The chamber has several observation windows which will allow for the experimental setup to be mounted outside (see figure 7b). Unfortunately there is no straight path through the chamber so the probe beam must be reflected back after passing through the sample by using a stationary mirror behind the sample holders. Preliminary measurements must be made to assure the viability of mounting

the mirror inside the chamber. New sample holders will have to be fabricated, leaving a window behind the substrate in order to let the probe beam pass through and reflect on the mirror. In this way an *in situ* absorption measurement can be achieved during the deposition process. This would be the first demonstration of *in situ* absorption loss measurements in a growth chamber.

To undertake development of this new prototype, preliminary measurements must be made inside the chamber to study the feasibility of mounting a fixed mirror on the back side of one the shutters. New sample holders must be designed, fabricated and tested. The rotation of each sample holder in the planetary system must be measured and thus estimate how often and how many times each sample passes in front of the fixed mirror. This will limit the temporal resolution in which the deposition of the IC can be characterized.

Due to the rotation of the samples, the pulsed pump system seems the most appropriate configuration. More detailed measurements with a pulsed pump laser should be performed using the setup already mounted on the optical table. It is crucial to understand the signal dependency with pulse duration. Bulk absorption of the substrate must be measured to quantify the sensitivity of the technique.

Most of the preliminary measurements will be performed by Carmen Menoni and Jorge Rocca's group with guidance from Oscar Martínez's group. In the meantime, further calculations and modeling will be performed to predict signal levels and design an adequate experimental configuration for the prototype. Once this stage is completed, Esteban Domené will travel to CSU to setup the new prototype. Preliminary measurements on known samples will be performed to align and calibrate the system adequately.

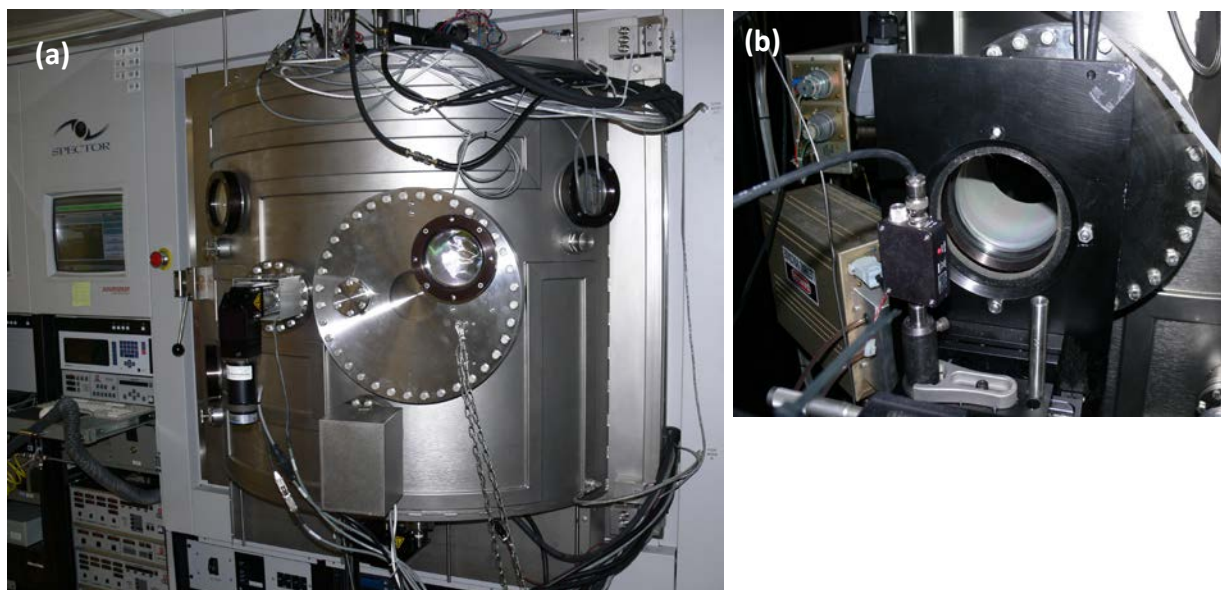


Figure 7. (a) Veeco Spector Ion Beam Sputtering machine. The chamber (on the right) with the electronics and control (on the left). (b) Back of the Veeco Spector Ion Beam Sputtering machine. One of the observation windows is shown. This is where the experimental setup will be mounted.

8. Publications and Products

“Thin film absorption loss characterization by focus error thermal lensing”, E. Domene, D. Schiltz, D. Patel, C.S. Menoni, J.J. Rocca and O.E. Martinez, Conference Abstract submitted to NINETEENTH SYMPOSIUM ON THERMOPHYSICAL PROPERTIES, Boulder, CO, USA, June 21 - 26, 2015.

“Thin film absorption loss characterization by focus error thermal lensing”, E. Domene, D. Schiltz, D. Patel, C.S. Menoni, J.J. Rocca and O.E. Martinez, manuscript in preparation, to be submitted to Optics Express, January 2015.

“Método Para Determinar La Absorción De Una Muestra Y Dispositivo Empleado En Dicho Metodo,” O.E. Martinez y E. Domene, Argentinian Patent Office, Filed December 15,m 2014.